



LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

LLNL-TR-663152

The Search for Electromagnetic Alteration of U-235m Decay

R. J. Casperson, R. O. Hughes, J. T. Burke, J. Dixon,
S. Fisher, J. Hamilton, R. Henderson, J. C. Robinson,
N. D. Scielzo, R. Soufli, E. L. Swanberg, E. B. Norman

October 23, 2014

Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States government. Neither the United States government nor Lawrence Livermore National Security, LLC, nor any of their employees makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States government or Lawrence Livermore National Security, LLC. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States government or Lawrence Livermore National Security, LLC, and shall not be used for advertising or product endorsement purposes.

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

The Search for Electromagnetic Alteration of ^{235m}U Decay

R. J. Casperson,* R. O. Hughes, J. T. Burke, J. A. Dixon, S. Fisher, J. Hamilton,
R. Henderson, J. C. Robinson, N. D. Scielzo, R. Soufli, and E. L. Swanberg
Lawrence Livermore National Laboratory, Livermore, California 94551, USA

E. B. Norman

Department of Nuclear Engineering, University of California, Berkeley, California 94720, USA

(Dated: October 27, 2014)

Electromagnetic modification of the ^{235m}U isomer has been tested by placing the decaying system in a number of different environments. The isomer decays by internal conversion, which involves the electromagnetic transfer of energy from the nucleus to an atomic electron, and it is expected that this process can be altered by changing the photon density of states available to the nucleus. The two approaches pursued during this work included direct detection of the electron produced by the isomer decay using a micro-channel plate detector, and detection of the scintillation light produced when the electron interacted with various scintillators. With direct electron detection, variations in the isomer population were observed before and after covering the isomer, but this effect is attributed to systematic errors associated with the implantation and covering process. With the scintillator approach, scintillation light from the decay of the isomer was measured for the first time, and the decay half-life was directly identified while the isomer was embedded in a bulk material. No modification was observed, which leads to the conclusion that internal conversion of ^{235m}U does not depend on the index of refraction of the surrounding environment in the way that spontaneous photon emission of an atomic system does. This result excludes many of the interesting applications of electromagnetic alteration of nuclear decay.

PACS numbers:

I. INTRODUCTION

Significant alteration of nuclear decay properties would have important consequences, ranging from applications, such as novel approaches to nuclear batteries [1], to physically interesting experiments [2, 3], where the half-life of the nuclear state plays an essential role in the interpretation. Quantum systems that decay by photon emission must couple to the electromagnetic modes of the local environment, and by modifying these modes, one can manipulate the rate of spontaneous emission [1, 4–7]. The decay of low-energy nuclear states typically involves gamma rays, which typically have $10^3 - 10^6$ times more energy than optical photons. At these energies, photon modes are very difficult to manipulate, and as a result, extremely low-energy nuclear transitions were the focus of this study.

The ^{235}U isotope has a long-lived nuclear state at an excitation energy of 76.5 eV [8], which is approximately thirty times larger than typical optical transitions. This is within the energy range where the electromagnetic modes can be substantially modified [4, 9], and is therefore a good test case. The decay of this state occurs by internal conversion, which involves the transfer of energy through virtual photons [10], which makes this case different from past studies of modified spontaneous emission in atomic systems.

There have been a number of studies focusing on the modification of the ^{235m}U decay from chemical effects [11–14], where the mechanism for modification is a change in the availability or energy of the atomic electrons present during internal conversion. This effect does not involve the index of refraction or density of photon modes of the local environment, and the largest observed magnitude was found to be 5%.

A later measurement was performed which focused on electromagnetic modification of ^{235m}U [15], and the measurement found a factor of ten half-life modification, which extends far beyond simple modified spontaneous emission theory [4, 9]. Repeat measurements using the same technique did not consistently reproduce the result [16], which calls into question the validity of the reported experimental technique, and whether internal conversion in nuclei can be modified in the same way that spontaneous photon emission in atomic systems can.

We have performed a number of measurement campaigns to study the effect that altering the surrounding electromagnetic environment has on the decay of ^{235m}U . The studied approaches ranged from indirect techniques, which involved measuring the decay rate before and after exposure to different environments, to direct approaches, where the half-life of the decay was measured continuously. The paper is structured in the following way: Section II describes the theoretical background of modified spontaneous emission and discusses the past experimental work studying spontaneous emission in more detail, Section III describes the indirect experimental measurements performed in this work, Section IV describes the direct experimental measurements, and Section V dis-

*Electronic address: casperson1@llnl.gov

cusses both sets of measurements.

II. BACKGROUND

Purcell predicted that the spontaneous emission rate of a decaying atomic state can be modified by placing the atom in a cavity with a similar size as the decay wavelength [17]. Spontaneous emission can be viewed as stimulated emission from vacuum-field fluctuations, and the resulting decay can be shown to directly depend on density of optical modes near the decaying system [6]. The optical mode density depends on the index of refraction of the material, but can be altered by creating a reflective cavity. Bjork showed that a planar one-dimensional mirror is adequate for significantly modifying the decay half-life of a system undergoing spontaneous photon emission [6].

The decay of an excited, low-energy nuclear state typically occurs via gamma-ray emission. For extremely low-energy transitions, a secondary process called internal conversion becomes more probable [10]. In this process, electromagnetic energy is transferred from the excited nucleus to an atomic electron, which can be described as an exchange of virtual photons. This process is thought to depend on density of local optical modes in the same way that spontaneous photon emission in atomic systems is known to [1], and one of the goals of this work was to test this hypothesis.

Most experimental studies of spontaneous emission modification have examined atomic systems, where electric dipole (E1) transitions are likely to occur. Many transition multiplicities are possible in nuclei, and the spontaneous emission rate should depend strongly on the multipolarity [4]. The index of refraction at x-ray and gamma-ray energies is typically less than unity, and can be calculated using atomic information [9]. The ^{235m}U isomer decays by an electric octupole (E3) transition [8], which has the following decay rate [4]:

$$W_{\text{solid}}^{EL} = f_L^2(\epsilon)\epsilon^{L-1/2}W_{\text{vacuum}}^{EL} \quad (1)$$

where L is multipolarity (e.g. 3 in the case of E3 transitions), ϵ is the permittivity, and f_L is a function representing the cavity model. There are two functions that can be used as the cavity model, and both give similar results for the decay of ^{235m}U :

$$f_L^{\text{real}} = \frac{\epsilon(2L+1)}{\epsilon(L+1)+L} \quad (2)$$

$$f_L^{\text{virtual}} = \frac{\epsilon L + L + 1}{2L + 1} \quad (3)$$

As mentioned in Section I, a past measurement was performed in which a large modification of the ^{235m}U decay half-life was observed [15]. The experimental technique involved depositing a layer of silver on a plate of

tungsten, and then collecting the ^{235m}U isomer on the silver surface, following the α -decay of ^{239}Pu . The decay rate was then measured, to establish the population of the ^{235m}U isomer. A second silver layer was deposited over the ^{235m}U , in order to form a solid material surrounding the isomer. After a fixed amount of time, the silver was boiled away, and the decay of the remaining ^{235m}U population was measured, to indirectly determine the half-life of the isomer while embedded in the silver.

One of the challenges in measuring the ^{235m}U decay is that the electron produced by internal conversion has a very short range of about 1 nm [18]. The consequence of this is that measuring the decay of the isomer while embedded deep within a material is extremely challenging, which causes indirect approaches to be more desirable. Unfortunately, a number of systematic errors can occur with indirect measurements, which make it difficult to establish the decay half-life while the isomer is embedded. Many of these effects will be discussed in Section III. Repeat measurements of ^{235m}U embedded in silver showed that the result was not reproducible [16], and a systematic error associated with indirect measurements is likely responsible.

Past work was performed in which the chemical effects of electron availability and shell energy on the ^{235m}U decay was studied [11–14]. These investigations showed correlations between the ^{235m}U half-life and the atomic shell structure, as well as with the oxidation state of the ^{235m}U compound. These results can be re-examined using a simple theory of modified spontaneous emission with the index of refraction of the surrounding material. The result is that on the surface of the material, the chemical shift must be much stronger than any potential electromagnetic shift. This is potentially due to the oscillatory nature of the spontaneous emission rate near an interface, and due to the depth at which the effect becomes large [19]. To properly study the electromagnetic effect of the ^{235m}U isomer decay, it is important to probe deeper than 4 nm in a material.

III. INDIRECT MEASUREMENT TECHNIQUES

A. Motivation

Directly measuring the ^{235m}U half-life deep within a material can be difficult, as the electron can only travel about 1 nm before it is unable to escape from the surface of the material [18]. This difficulty can be overcome by counting the isomer population before and after placing it in a bulk material, and by using these populations to identify any deviation from the ^{235m}U vacuum half-life of 26 minutes. The approach initially proposed for this work was to collect the isomer using an Argon buffer gas, and to press the wafer the isomer was collected on against a second wafer. It was quickly established that with standard clean room conditions, dust will prevent the wafers from sticking together and acting as a bulk

TABLE I: Predicted half-lives of the ^{235m}U isomer in the materials used for indirect measurements, using a simple theory for modified spontaneous emission [4, 9].

Material	Density (g/cm ³)	$ \epsilon $	Half-life (min)	Modification (%)
Platinum	21.45	0.78	66	153
Mercury	13.53	0.85	47	81
Quartz	2.65	0.92	35	33
Fused silica	2.2	0.93	33	26
Methanol	0.79	0.95	31	18
Ice	0.92	0.95	31	21

solid.

A modified approach involved collecting the isomer on the surface of a wafer, and covering the wafer with a liquid, to simulate the isomer being embedded in a bulk material. As will be shown, residue from the liquids lead to losses, so an additional approach was used, which involved cooling the wafer until it condensed moisture from the air, to simulate a bulk material, and returning the wafer to the vacuum system before the ice melted. A final measurement was made using molybdenum/silicon (MoSi) multilayer mirrors, to investigate the effect of photonics materials that were tuned to the 16.2 nm ^{235m}U transition wavelength.

A number of different materials and interfaces were used during these measurements, and rough estimates for the potential electromagnetic modification can be made using calculated x-ray indices of refraction [9] and a simple theory of modified spontaneous emission for E3 decays [4]. A table of the relevant materials for this section can be seen in Table I.

B. Experimental setup

Counting single electrons is most practical in a vacuum system, using an electron multiplier to create a large signal from individual electrons. For the measurements performed in this work, an 18" diameter by 9" tall vacuum chamber was pumped down to pressures of 10^{-6} Torr using an Edwards xds5 scroll pump and a Pfeiffer TPH330 turbo pump. An aluminum source can was machined to allow for the collimation of a 2- μCi ^{239}Pu source, which was electroplated to a stainless steel plate and heat treated. A gas flow system was constructed using an MKS type 649 pressure controller, and several vacuum valves. Argon gas was provided by a high-pressure gas cylinder and a pressure regulator.

A McLennan MRIG11S translation arm was used to transport a catcher plate between the ^{239}Pu source can and the 1" diameter, Photonis micro-channel plate electron detector. A solid region of the arm blocked the source can when the micro-channel plate was counting the isomer, in order to block any electrons produced by the ^{239}Pu α -decay. A high-voltage electrode was con-

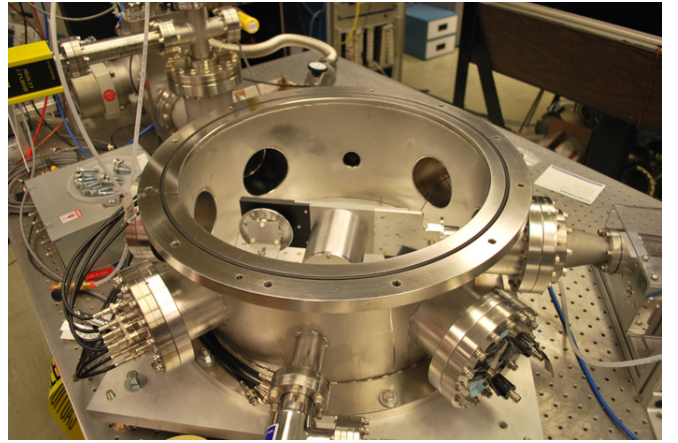


FIG. 1: A photo of the vacuum system used to perform indirect measurements by counting electrons. The disk-like object on the left of the chamber is the microchannel plate detector, and the large aluminum cylinder in the middle of the chamber is the source can. The long rectangular object in the vacuum chamber is the translation arm.

nected to the catcher, to drift the isomer through the Argon gas and onto the surface of the catcher. The high-voltage was also used to accelerate the electrons produced by the isomer decay into the MCP, which increased the collection efficiency. A photo of the vacuum system can be seen in Fig. 1.

The data was recorded with a VME-based data acquisition system, which included an XVB602 VME CPU, and an MADC-32 peak-sensing ADC. A fast electronics branch provided the trigger and time data, and a slow branch provided the shaped signal for the peak-sensing ADC. When analyzing the data, a gate was placed on the single electron amplitude spectrum in order to cut out electrical noise.

C. Results

A number of different materials and conditions were tested with the electron counting setup, and most gave a half-life very similar to the ^{235m}U vacuum value of 26 minutes [8]. Four different material combinations were tested in the indirect measurement phase of this work: platinum/mercury, quartz/methanol, fused silica/ice, and molybdenum/silicon.

1. Platinum and mercury

The first modification to the original strategy of pressing wafers together was to dip one wafer in mercury, which is a metallic liquid with a density of 13.5 g/cm³. The goal of this approach was to create a dense layer of metal that conformed to the surface of the catcher wafer, regardless of any dust that was present.

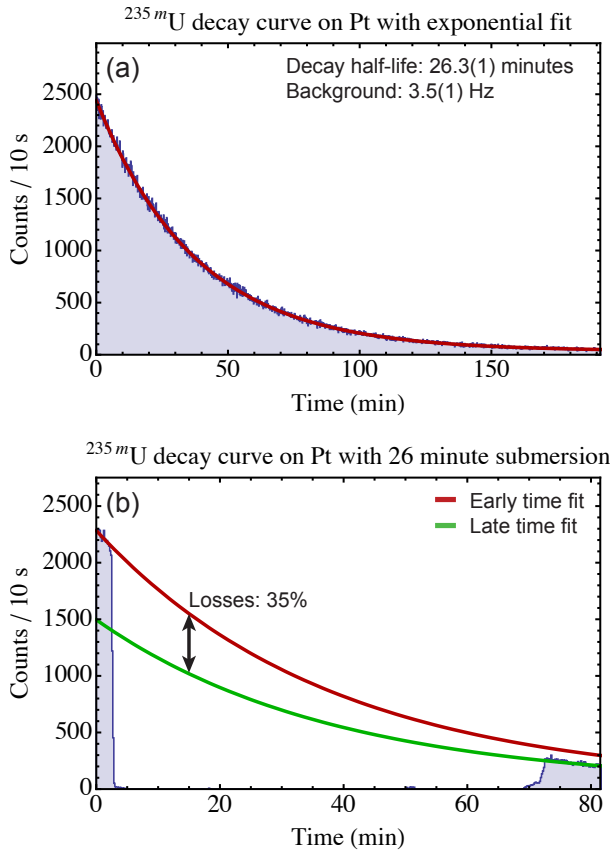


FIG. 2: a) Full decay curve of ^{235m}U on platinum, after collecting the isomer in argon. b) Decay curves of ^{235m}U on platinum, before and after submersion in mercury, after collecting the isomer in argon. 35% losses were observed after submersion in mercury.

Mercury forms an amalgam with most metals, and wafers that include such metals would dissolve when dipped in mercury. To prevent this, silicon wafers with 200 nm platinum coatings were used to collect the ^{235m}U isomer. To establish the reference half-life for the mercury submersion, a baseline measurement was first made by collecting the isomer on the platinum wafer, and counting the full decay curve. This decay curve can be seen in Fig. 2a, which shows a half-life of 26.3(1) minutes.

The mercury dip measurement involved pushing the wafer down into a small container of mercury, shallow enough that the surface of the liquid was not broken. A photo of a platinum wafer before submersion in mercury can be seen in Fig. 3. Several repeat measurements with this approach were made, and one example can be seen in Fig. 2b. In most cases, losses in the ^{235m}U population were observed, which is consistent with either the isomer dissolving in the mercury, or a layer of residue forming on the platinum coating.

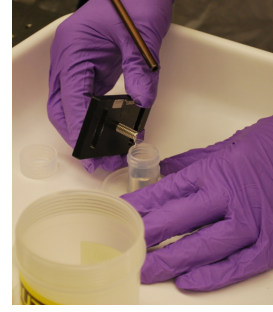


FIG. 3: A photo of the platinum coated wafer, before submersion in a small container of mercury. The platinum wafer is mounted on a stainless steel bolt, which is attached to a metal plate.

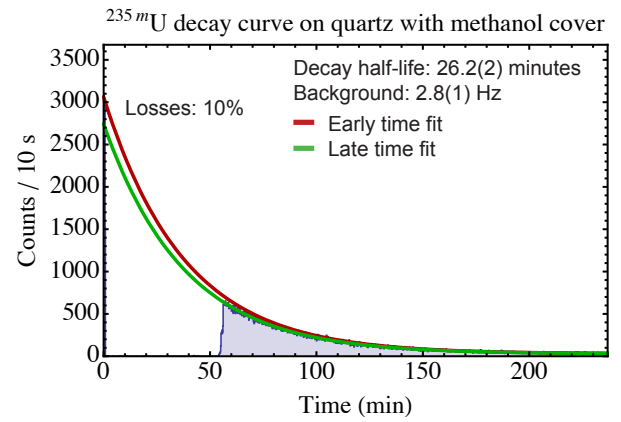


FIG. 4: Decay curves of ^{235m}U on quartz, before and after being covered with methanol, after collecting the isomer in argon. 10% losses of the isomer were observed after covering the wafer with methanol.

2. Quartz and methanol

During the platinum and mercury measurements, it was unclear whether platinum resisted forming an amalgam at the nanometer scale, and whether the uranium would remain stuck to the surface after exposure to the mercury. One strategy that reduces the risk of lost isomer is to cover the wafer with a liquid that is eventually evaporated. The specific case that was tested in this work was that of a quartz wafer, covered with methanol. A sapphire wafer was placed on top of the methanol to reduce the evaporation rate, and was removed after the desired amount of time. Fig. 4 shows the result of this measurement.

After the methanol evaporated, a 10% loss in the isomer population was observed. This is most likely due to non-volatile residues in the methanol covering the isomer, and preventing the decay electrons from escaping from the wafer surface.

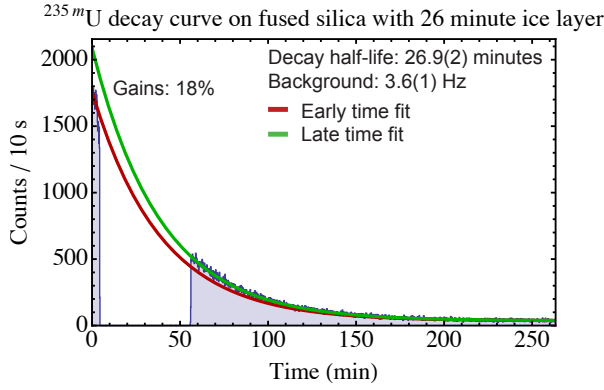


FIG. 5: Decay curves of ^{235m}U on fused silica, before and after being covered with a layer of ice for about 26 minutes, after collecting the isomer at vacuum. 18% gains of the isomer were observed after covering the wafer with a layer of ice.

3. Fused silica and ice

One of the potential sources of isomer loss in the platinum/mercury measurement was exposure of the isomer itself to the mercury. In the quartz/methanol measurement a residue may have formed, preventing the electrons from escaping from the surface. To address both of these problems, isomer was collected at vacuum, and the wafer was cooled to a temperature where moisture condensed from the air. By collecting the isomer at vacuum, the 88 keV ^{235m}U recoils from ^{239}Pu α decay implant several nanometers deep within the wafer. Only isomer near the surface will be detectable, but the implantation process does allow for detectable isomer that is not immediately on the surface of the wafer.

Cooling the wafer was accomplished by placing it on top of powdered dry ice (i.e. solid CO_2), which has a temperature of -78.5°C . After about one minute, the wafer temperature fell below 0°C , and a thin layer of condensation froze to the surface of the wafer. The wafer was kept in the container of dry ice for a set amount of time, and was then placed back in the vacuum system. The ice then sublimated, and electrons were counted from the surface of the wafer.

Fig. 5 shows the decay curve before and after freezing a layer of ice on the wafer for 26 minutes. The 18% gain shown in the figure would correspond to an extended half-life, if the cause of the increased rate was electromagnetic modification of spontaneous emission. There are a number of other possibilities, including isomer migration, crystal annealing after exposure to air, and a change in the oxidation state after exposure to air.

Table II shows the amount of gain for a variety of exposure times and time delays between early and late time fits. With minimal exposure time, a gain of between 6% and 12% was found, indicating a change in the detection efficiency that is independent of the ice coverage. The longer exposure times showed larger gains, which

TABLE II: Measured gain in the ^{235m}U isomer population after exposure to a thin layer of ice. The exposure time is a rough estimate, as tracking the ice coverage precisely was not possible. The amount of time between early and late time fits is listed.

Exposure (min)	Delay (min)	Gain (%)
0	24	6
0	81	12
5	28	10
10	29	7
26	57	20
52	85	30
52	82	-4

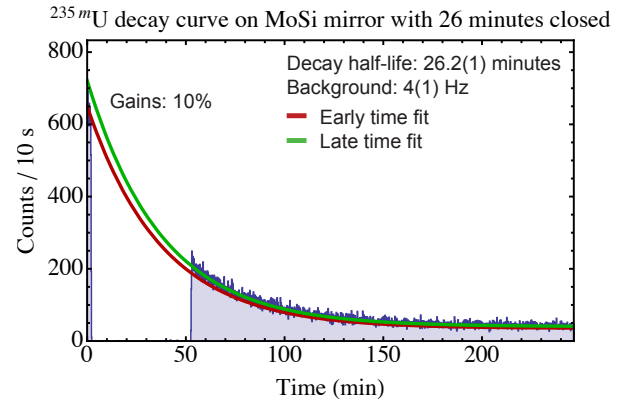


FIG. 6: Decay curves of ^{235m}U on a MoSi multilayer mirror, before and after being pressed against a second MoSi mirror for 26 minutes, after collecting the isomer at vacuum. 10% gains of the isomer were observed after separating the two mirrors.

could correspond to a longer half-life, but repeat measurements gave inconsistent results. The ice exposure had the largest population changes found during this work, and they illustrate the challenge of isolating efficiency effects during indirect measurements.

4. Molybdenum and silicon multilayer mirrors

One of the original goals for this project was to create multilayer mirrors that are highly reflective at 76.5 eV, which is the transition energy of interest [8]. These mirrors were then to be used to create a photonic band-gap, by pressing two mirrors together. The multilayer mirrors were constructed using magnetron sputtering, with 3.4 nm thick molybdenum layers and 5.1 nm thick silicon layers. The reflectivity of these mirrors was measured using the Advanced Light Source at Lawrence Berkeley National Laboratory. The mirrors were measured to have a reflectivity of 52% at normal incidence for the 16.2 nm wavelength of interest.

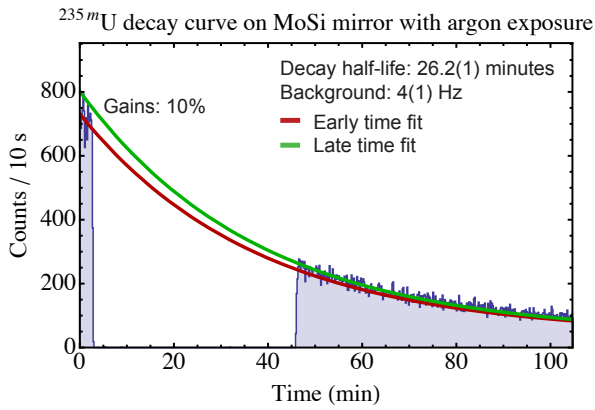


FIG. 7: Decay curves of ^{235m}U on molybdenum/silicon (MoSi) multilayer mirror, before and after exposing the mirror to argon, after collecting the isomer at vacuum. 10% gains of the isomer were observed after exposing to argon.

Fig. 6 shows a decay curve before and after two MoSi multilayer mirrors were pressed together, after collecting the isomer at vacuum on one of the mirrors. The 10% gains found were similar to the gains found with fused silica after exposure to atmosphere. To test whether pressing the mirrors together was responsible for the increased rate, data was recorded with the same exposure time to argon, without pressing the mirrors together. Fig. 6 shows a decay curve illustrating this, and the same gains were found without pressing the mirrors together. This again shows the difficulties of identifying a half-life modification using an indirect measurement technique. Additional measurements were made to better understand the origin on these gains, and it was found that they only occur when the isomer is collected at vacuum.

IV. DIRECT MEASUREMENT TECHNIQUES

A. Motivation

Many of the systematic errors that result from indirect measurements can be eliminated by directly measuring the decay curve. The challenge with direct measurements is that the electrons produced in a bulk material cannot be directly counted. One approach that would allow for remote detection of the decay is to measure the light produced by the electron interacting with a scintillator. Such measurements have been attempted in the past [20], and scintillation light from this process has never been identified. One of the main challenges with counting ^{235m}U specifically is that the isomer recoils and α -particles produced by ^{239}Pu α -decay causes a substantial amount of damage to the scintillator crystal [21], potentially resulting in after-glow and altered scintillation properties.

Many different scintillator crystals were used during these measurements, and rough estimates for the potential electromagnetic modification can again be made us-

TABLE III: Predicted half-lives of the ^{235m}U isomer in the materials used for direct measurements, using a simple theory for modified spontaneous emission [4, 9].

Material	Density (g/cm ³)	$ \epsilon $	Half-life (min)	Modification (%)
$\text{Bi}_4\text{Ge}_3\text{O}_{12}$	7.13	0.90	38	45
CdWO_4	7.90	0.85	46	77

ing calculated x-ray indices of refraction [9] and a simple theory of modified spontaneous emission for E3 decays [4]. A table of the two most promising materials can be seen in Table III.

B. Experimental setup

For this series of measurements an Electron Tubes Type 9426 photomultiplier tube (PMT), with a wavelength range of 210-630 nm was used to detect single photons. At room temperature, the photocathode of the PMT produces a large rate of thermal electrons, which are indistinguishable from the photons of interest. Using a photocathode that only responds to shorter wavelengths improves this dark count rate, and cooling the PMT improves the rate dramatically. The PMT used for these measurements was cooled to -18°C using a Thermo Fisher Scientific chiller, which reduced the dark count rate from 1000 Hz to about 10 Hz. The chiller used a 50:50 mix of ethylene glycol and water.

The same aluminum source can shown in Section III was used to contain the ^{239}Pu source. Scintillators can have a number of backgrounds with long decay constants, which made it essential to perform background measurements using a ^{240}Pu source. The ^{240}Pu source produces energetically similar recoils and α particles as the ^{240}Pu source, but does not produce the ^{235m}U isomer.

The ^{235m}U isomer was collected on a wafer, which was then transferred between the source can and the PMT using a manual translation arm. Two tungsten plates were placed between the source can and the PMT, to prevent light and x-rays from reaching the scintillator and the PMT. A photo of the vacuum system where the photon counting measurements were performed can be seen in Fig. 8.

C. Results

A number of scintillating crystals were tested in the direct measurement phase of this project, and most materials had substantial afterglow, which prevented the identification of the ^{235m}U isomer decay. Bismuth germanate (BGO) has the chemical formula $\text{Bi}_4\text{Ge}_3\text{O}_{12}$, and is predicted to have a large half-life modification, which can be seen in Table III. It has a density of 7.13 g/cm³, and a modest scintillation light yield of 8000 photons/MeV [22].

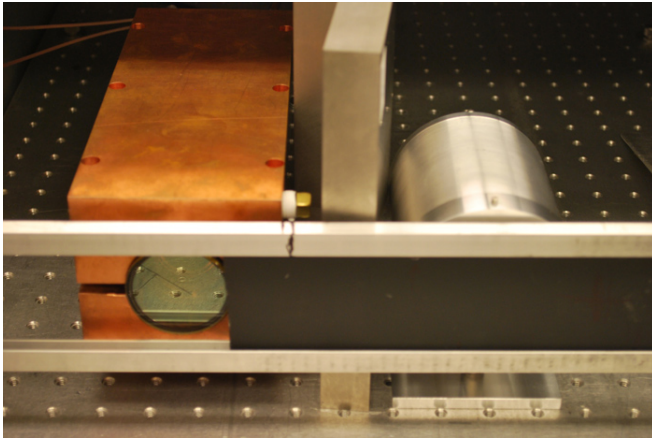


FIG. 8: A photo of the vacuum system used to perform direct measurements by counting photons. The reflective circle on the left of the chamber is the PMT, which was surrounded by a large, chilled, copper block. The two vertical plates are tungsten, and the aluminum cylinder on the right is the source can. The long flat object in the foreground is the manual translation arm.

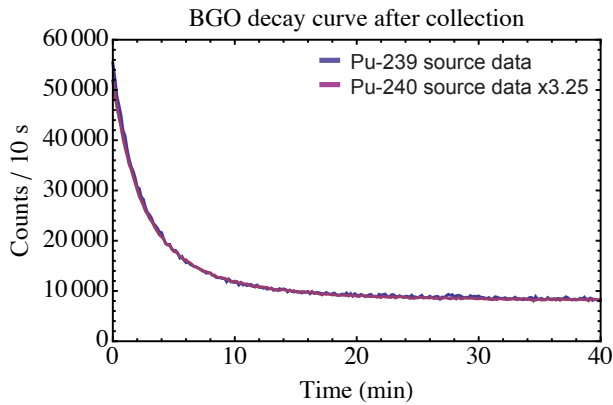


FIG. 9: Light production in the BGO crystal after uranium recoil implantation from ^{239}Pu and ^{240}Pu sources. The ^{240}Pu data has been scaled up by a factor of 3.25 for comparison.

Scintillation light yields are known to depend strongly on electron energy [23], and extrapolating down to the ^{235m}U decay energy can be difficult. The time decay constant of BGO is about 300 ns.

Fig. 9 shows a comparison of the decay curve after uranium recoil implantation when using ^{239}Pu and ^{240}Pu sources. The decay curves are nearly identical for the two source exposures, and the produced light is attributed to α -induced afterglow. This shows that BGO is not an appropriate material for measuring the ^{235}U isomer decay.

Cadmium tungstate, which has the chemical formula CdWO_4 , was also tested as a scintillator. It has a density of 7.9 g/cm^3 , and a higher light yield of 15000 photons/MeV [24]. It also has good radiation resistance, and

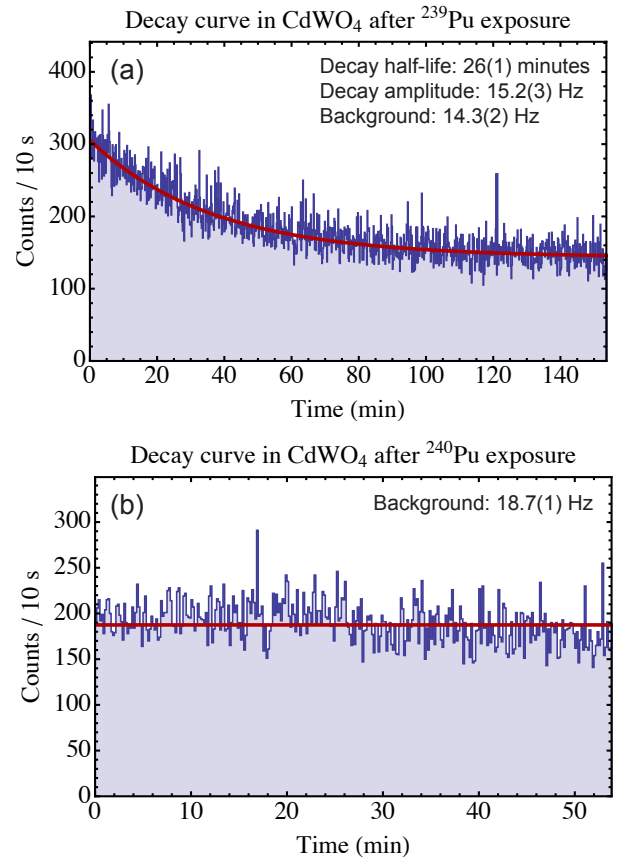


FIG. 10: Light production in the CdWO_4 crystal after uranium recoil implantation from a) ^{239}Pu and b) ^{240}Pu sources. The ^{239}Pu data shows the vacuum ^{235m}U decay half-life of 26(1) minutes.

a time decay constant of $10 \mu\text{s}$, which is unusually long among scintillators. Fig. 10 shows the decay curves in CdWO_4 after exposure to ^{239}Pu and ^{240}Pu sources. The ^{240}Pu source data shows a relatively flat background, and the ^{239}Pu source data shows a strong decay curve, corresponding to a 26(1) minute half-life.

V. DISCUSSION

A number of different indirect measurement techniques were used in this work to identify whether the ^{235m}U half-life would change when the isomer is placed in unique electromagnetic environments. Each measurement showed a change in the isomer population, which illustrates the difficulty in interpreting potential half-life modifications with indirect measurements. For measurements where the isomer was collected in argon, and the wafer was exposed to a liquid, losses were observed. This most likely resulted from isomer dissolving, or being covered by non-volatile residues from the liquid. For measurements where the isomer was collected in vacuum, and was then exposed to air or ice, the increased rate likely re-

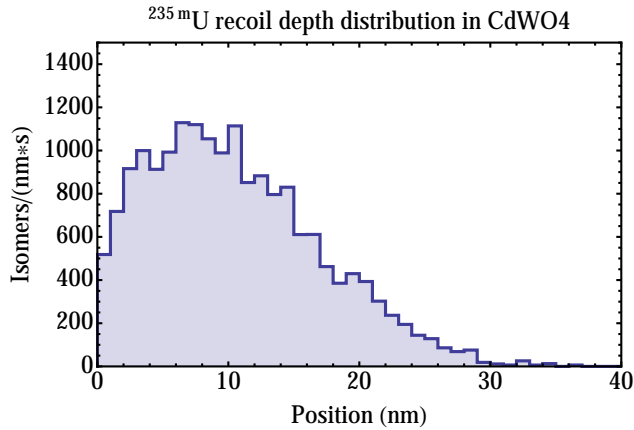


FIG. 11: Depth distribution of ^{235m}U recoils in the CdWO_4 crystal, as calculated in SRIM.

sulted from annealing of the damaged crystal, migration of the isomer in the crystal, or oxidation of the uranium isomer.

The past measurements of ^{235m}U embedded in silver used the indirect measurement technique, and showed consistent gains that did not appear to depend on exposure time in a statistically significant way [15, 16]. Given the many sources of systematic errors for indirect measurements, it seems unlikely that the spontaneous emission rate was modified electromagnetically, as was originally suggested in the first reported result [15].

Many scintillators have been tested for direct measurement of scintillation light produced by the isomer decay electrons, in order to avoid the problems that occur in indirect measurements. Most scintillators had substantial afterglow, which interfered with the detection of the isomer decay. After collecting the ^{235m}U recoils in the scintillator CdWO_4 , a decay curve of 26(1) minutes was detected, which is consistent with the known vacuum half-life of ^{235m}U . This is the first time that scintillation light from the decay of ^{235m}U has been measured and identified.

The recoil depth distribution of the ^{235m}U recoils implanted in CdWO_4 can be calculated in SRIM [21], by entering the material properties and generating appropriate initial conditions in the calculation, such as a depth and direction. The resulting trajectories can be analyzed to enforce the geometric conditions of the experimental setup. The results of such a calculation can be seen in Fig. 11. The distribution peaks at 8 nm, and extends up to 30 nm deep. Although interface effects are known to occur for modified spontaneous emission [19], they are

not expected to inhibit the effect for ^{235m}U beyond a depth of about 4 nm.

As shown in Table III, the simple modified spontaneous emission theory would give ^{235m}U isomer a half-life of 46 minutes in CdWO_4 , which would be a 77% suppression of the decay. The lack of any measurable modification of the ^{235m}U half-life, given the deep distribution of isomer shown in Fig. 11, leads to the conclusion that internal conversion of ^{235m}U is not affected by the index of the refraction of the surrounding environment in the same way as spontaneous photon emission in atomic systems.

VI. CONCLUSIONS

Electromagnetic modification of the ^{235m}U isomer has been tested by measuring the decay of the isomer in a number of different environments. The isomer decays by internal conversion, which involves the transfer of energy from the nucleus to an atomic electron through virtual photon exchange, and it was expected that this transfer of energy could be altered by changing the photon density of states available to the nucleus. Several indirect measurements were performed, in which the isomer population was measured before and after embedding it within different environments. All indirect measurements showed population losses or gains, even when the isomer was not covered. To mitigate the systematic errors inherent in indirect measurements, several direct measurements of the isomer half-life in unique environments were performed. The isomer was embedded in scintillating crystals, and the scintillation light produced by the isomer decay was measured for the first time. The measured isomer half-life was found to match the vacuum half-life, which is not consistent with the simple theory of modified spontaneous emission for internal conversion in ^{235m}U . This result excludes many of the interesting applications of electromagnetic alteration of nuclear decay.

VII. ACKNOWLEDGMENTS

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. The Advanced Light Source is supported by the Director, Office of Science, Office of Basic Energy Sciences, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231.

-
- [1] R. W. Ziolkowski and D. M. Gogny, Progress In Electromagnetics Research **18**, 285 (1998).
 - [2] A. D'Eer, C. Wagemans, M. Nève de Mévergnies, F. Gönnenwein, P. Geltenbort, M. S. Moore, and

J. Pauwels, Phys. Rev. C **38**, 1270 (1988), URL <http://link.aps.org/doi/10.1103/PhysRevC.38.1270>.

- [3] W. Younes and H. C. Britt, Phys. Rev. C **67**, 024610 (2003), URL <http://link.aps.org/doi/10.1103/PhysRevC.67.024610>.

- 1103/PhysRevC.67.024610.
- [4] E. Tkalya, Journal of Experimental and Theoretical Physics **92**, 61 (2001), ISSN 1063-7761, URL <http://dx.doi.org/10.1134/1.1348461>.
 - [5] H. T. Dung, S. Y. Buhmann, and D.-G. Welsch, Phys. Rev. A **74**, 023803 (2006), URL <http://link.aps.org/doi/10.1103/PhysRevA.74.023803>.
 - [6] G. Björk, S. Machida, Y. Yamamoto, and K. Igeta, Phys. Rev. A **44**, 669 (1991), URL <http://link.aps.org/doi/10.1103/PhysRevA.44.669>.
 - [7] G. L. J. A. Rikken, Phys. Rev. A **51**, 4906 (1995), URL <http://link.aps.org/doi/10.1103/PhysRevA.51.4906>.
 - [8] E. Browne, Nuclear Data Sheets **98**, 665 (2003), ISSN 0090-3752, URL <http://www.sciencedirect.com/science/article/pii/S0090375203900056>.
 - [9] B. Henke, E. Gullikson, and J. Davis, Atomic Data and Nuclear Data Tables **54**, 181 (1993), ISSN 0092-640X, URL <http://www.sciencedirect.com/science/article/pii/S0092640X83710132>.
 - [10] N. Tralli and G. Goertzel, Phys. Rev. **83**, 399 (1951), URL <http://link.aps.org/doi/10.1103/PhysRev.83.399>.
 - [11] H. Mazaki and S. Shimizu, Phys. Rev. **148**, 1161 (1966), URL <http://link.aps.org/doi/10.1103/PhysRev.148.1161>.
 - [12] M. N. de Mévergnies, Phys. Rev. Lett. **23**, 422 (1969), URL <http://link.aps.org/doi/10.1103/PhysRevLett.23.422>.
 - [13] M. N. de Mevergnies, Phys. Rev. Lett. **29**, 1188 (1972), URL <http://link.aps.org/doi/10.1103/PhysRevLett.29.1188>.
 - [14] M. de Mevergnies and P. D. Marmol, Physics Letters B **49**, 428 (1974), ISSN 0370-2693, URL <http://www.sciencedirect.com/science/article/pii/0370269374906261>.
 - [15] V. V. Koltsov and A. A. Rimsky-Korsakov, Bull. Acad. Sci. USSR, Phys. Ser. **53**, 21 (1989).
 - [16] O. V. Voryhalov, E. A. Zaitsev, V. V. Koltsov, and A. A. Rimsky-Korsakov, Bull. Rus. Acad. Sci. Phys. **56**, 81 (1992).
 - [17] E. M. Purcell, Phys. Rev. **69**, 681 (1946).
 - [18] H. Shinotsuka, S. Tanuma, C. Powell, and D. Penn, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms **270**, 75 (2012), ISSN 0168-583X, URL <http://www.sciencedirect.com/science/article/pii/S0168583X11008809>.
 - [19] H. Khosravi and R. Loudon, Proc. R. Soc. Lond. A **433**, 337 (1991).
 - [20] E. Swanberg, Ph.D. thesis, UC Berkeley: Nuclear Engineering (2012).
 - [21] J. Ziegler, M. Ziegler, and J. Biersack, Nuclear Instruments and Methods in Physics Research, Section B: Beam Interactions with Materials and Atoms **268**, 1818 (2010), cited By (since 1996)608, URL <http://www.scopus.com/inward/record.url?eid=2-s2.0-77953138323&partnerID=40&md5=2770d3e9054108ba976b98b23a5485b1>.
 - [22] J. A. Mares, A. Beitlerova, M. Nikl, N. Solovieva, C. D'Ambrosio, K. Blazek, P. Maly, K. Nejezchle, and F. de Notaristefani, Radiation Measurements **38**, 353 (2004), ISSN 1350-4487, proceedings of the 5th European Conference on Luminescent Detectors and Transformers of Ionizing Radiation (LUMDETR 2003), URL <http://www.sciencedirect.com/science/article/pii/S1350448704001234>.
 - [23] I. V. Khodyuk, P. A. Rodnyi, and P. Dorenbos, Journal of Applied Physics **107**, 113513 (2010), URL <http://scitation.aip.org/content/aip/journal/jap/107/11/10.1063/1.3431009>.
 - [24] I. Holl, E. Lorenz, and G. Mageras, Nuclear Science, IEEE Transactions on **35**, 105 (1988), ISSN 0018-9499.